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ACID GAS REMOVAL CHARACTERISTICS OF CORONA RADICAL SHOWER SYSTEM FOR A TREATMENT OF STATIONARY ENGINE FLUE GAS

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ABSTRACT

Acid gas removal experiments are carried out in large bench scale corona radical shower reactor. A simulated stationary engine flue gas is air mixed with NO, SO_2 and CH_4. Optimums for acid gas removal rate have been conducted in terms of the ammonia to acid gas molar ratio, the applied voltage and the additional gas velocity. Both the removal efficiencies of NO and SO_2 increase with increasing applied voltage and decreasing initial NO concentration. It is also shown that high concentrations of CH_4 have a negative effect on NO removal, where part of the NO converted only to NO_2 but not to ammonium nitrate aerosol particles.

INTRODUCTION

NO_x and SO_x are one of air pollutants and the major cause of acid rain. Many NO_x and SO_x conversion techniques such as wet scrubber, selective catalytic reactor, sorbent injection, low NO_x burner, etc., have been used. More recently, non-thermal plasma techniques become a commercial plant, however, the energy efficiency of the non-thermal plasma reactors have not been optimum yet. For example, the electron beam, barrier discharge and pulsed corona reactors, i.e. direct plasma treatments of flue gases, may loss input energy to activate unwanted components of flue gases such as CO_2, N_2, etc. Hence, the corona discharge radical injection techniques have been developed. On the other hand, the treatments of an engine combustion flue gas by non-thermal plasmas have been investigated by many researchers [1-5]. In this work, an experimental investigation has been conducted to remove NO_x and SO_2 from the stationary engine flue gases.

EXPERIMENTAL APPARATUS

The schematics of experimental test loop and corona radical shower electrode are shown in Fig. 1. In stationary engine flue gas treatments, the combustion flue gas was normally diluted

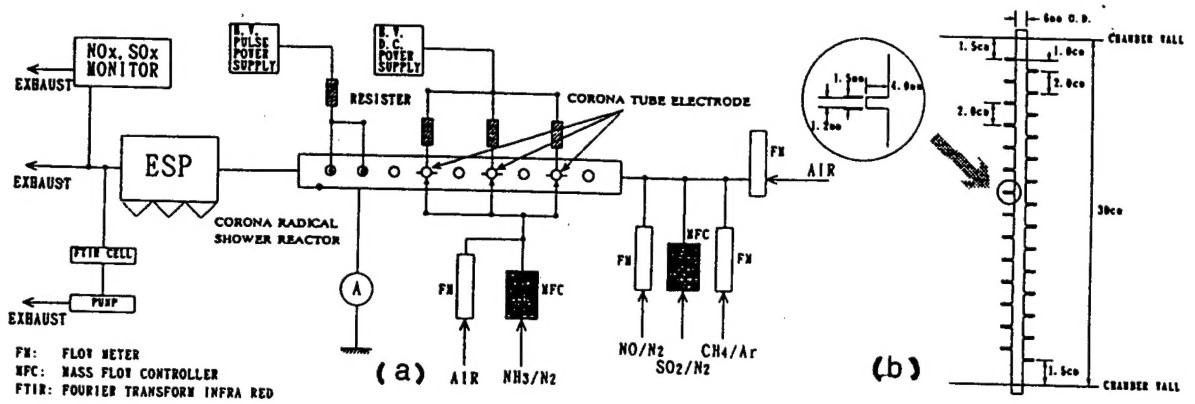

Fig. 1 Schematics of experimental flow loop (a) and corona radical shower electrode (b).

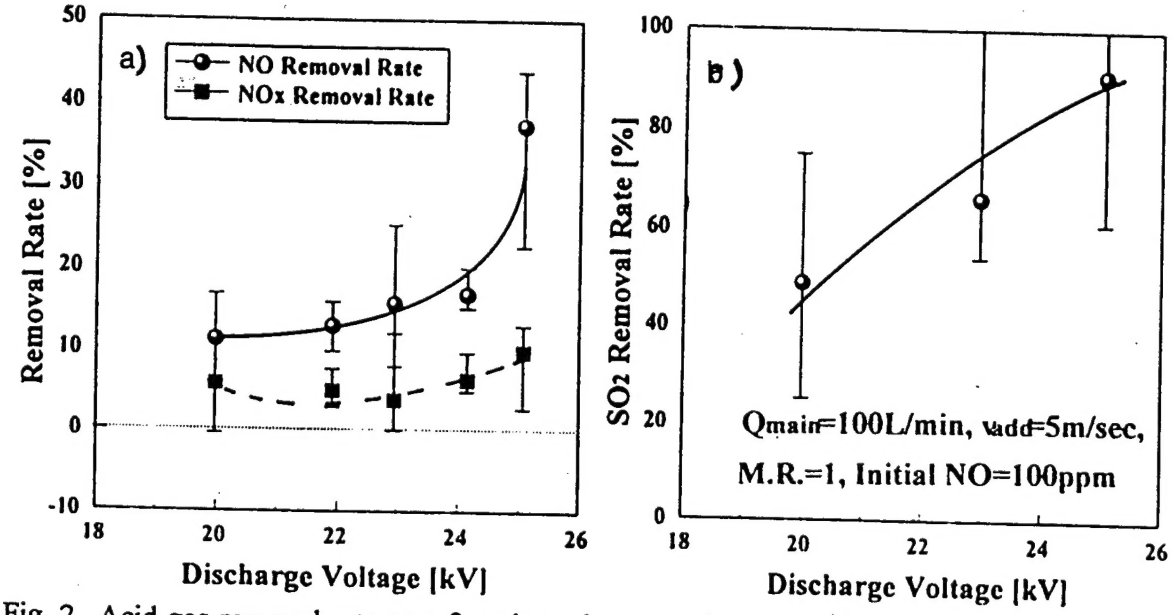

Fig. 2 Acid gas removal rate as a function of corona shower system applied voltage V, where bar in the each experimental points indicated minimum and maximum values. a) NO and NO_x and b) SO_2.

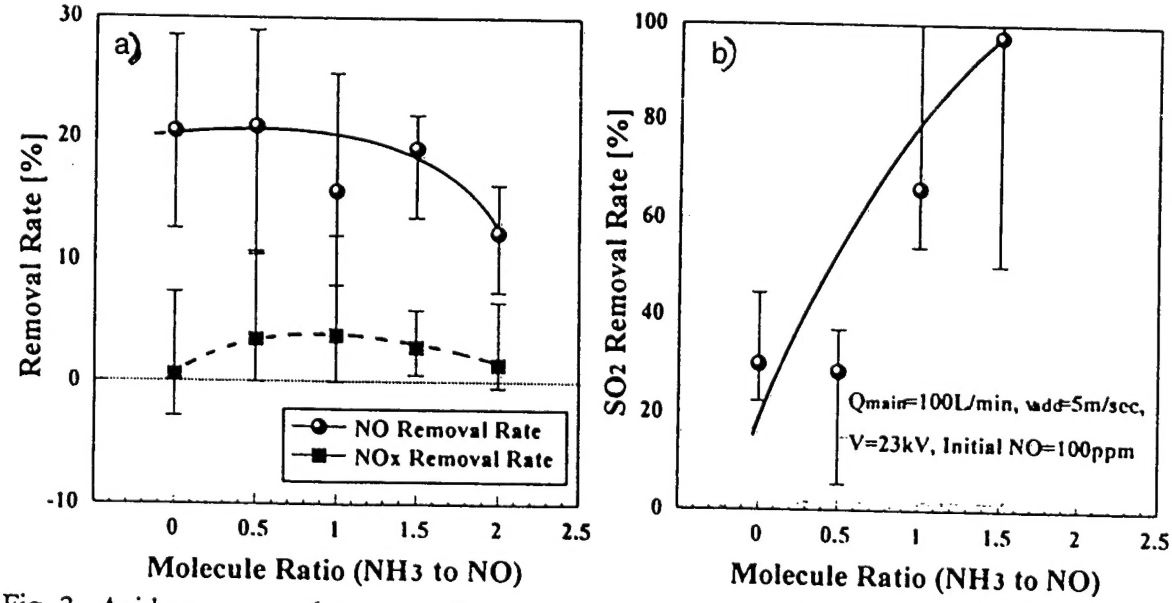

Fig. 3 Acid gas removal rate as a function of ammonia to acid gas molecule ratio MR. a) NO and NO_x and b) SO_2.

by the air for cooling down to room temperature. In the present simulated flue gas test, the concentration ratio between NO and SO_2 or CH_4 are fixed at 0.128 or 1.667, respectively, and then NO initial concentration was studied from 30 to 100 ppm. NO, NO_2 and SO_2 concentrations were measured by the Green Line gas analyzer and the trace by-products are determined by the Fourier Transform Infra-Red spectroscopy (FTIR). The aerosol particles generated by the acid gases and ammonia/methane related plasma processes were collected by the electrostatic precipitator operated at -19 kV dc at the downstream of the reactor. The size of the reactor is (10×30×100 cm) and three radical injectors are placed in series a shown in Fig. 1. The corona radical shower electrode used was 6 mm o.d. tube equipped with 28 hollow electrodes (1.2 mm i.d./1.5 mm o.d.) as shown in Fig. 1b. Additional gas consisting of air-NH_3 mixture was injected from these hollow electrodes to the reactor via corona discharge generated by a positive dc high voltage at the edge of the hollow electrodes.

EXPERIMENTAL RESULTS

NO, NO_x and SO_2 removal rate as a function of applied voltage, ammonia-to-acid gas molecule ratio MR, additional gas superficial velocity and NO initial concentration are shown in Figs. 2, 3, 4 and 5, respectively, where the bar for each experimental point shows minimum and maximum value observed during the time transient tests. NO, NO_x and SO_2 removal rate increases with increasing NO initial concentration in flue gases as shown in Figs. 2 and 5, respectively, as expected from radical chemistry and heterogeneous reactions [5]. However, acid gas removal rate has an optimum values in terms of MR and corona radical shower system additional gas velocity as shown in Figs. 3 and 4, respectively, where NO and NO_x removal rate is maximum near molecule ratio between 1 to 1.5 and additional gas velocity near 8 m/s. Based on analyses of Ohkubo et al [6], the effect of additional gas velocity can be explained by the better mixing of ammonia radicals to react with acid gases in lower velocity regions, where the mixing effect increases with increasing additional gas flow rates. For higher additional gas velocities, the ammonia radicals ejected from the hollow electrode may have shorter residence time in the reactor and only generate narrow flow channels to reduce reactions. Hence, the optimum additional gas flow velocity exists for the present range of flue gas flow rate (6 Nm^3/h or reactor superficial velocity of 0.33 m/s).

Based on corona discharge gas phase and heterogeneous reactions with ammonia [5], the SO_2 removal rate increases with increasing MR as shown in Fig. 3. However, if we examined discharge by-products, ammonia and methane relative concentrations ($\propto$ absorbance of IR) as shown in Fig. 6, CH_4 and N_2O concentration become minimum at molecule ratio near 1 to 1.5 where NH_3, H_2O and ϕ-CHO were determined by the IR absorptions at the wave length

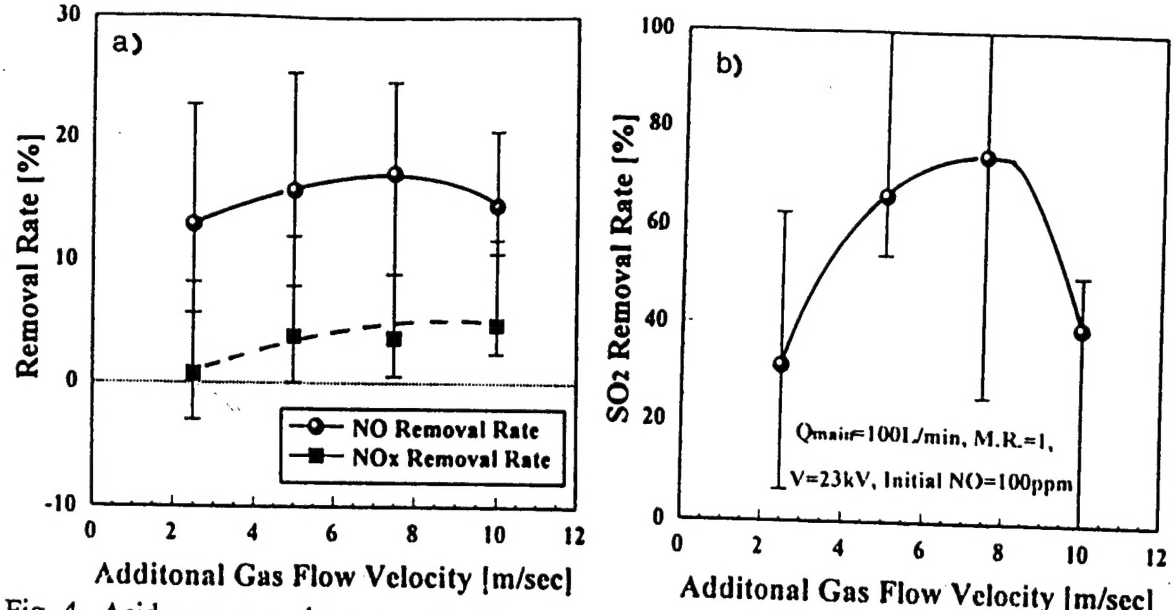

Fig. 4 Acid gas removal rate as a function of corona shower system hollow electrode additional gas flow velocity V_{add}. a) NO and NO_x and b) SO_2.

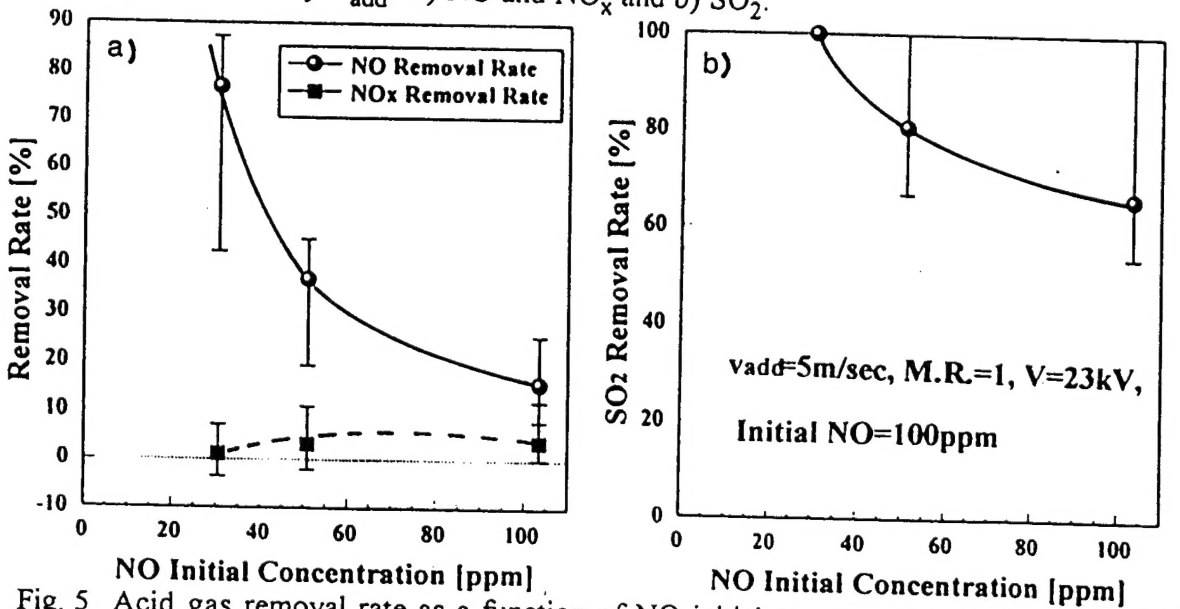

Fig. 5 Acid gas removal rate as a function of NO initial concentration in flue gases $[NO]_o$. a) NO and NO_x and b) SO_2.

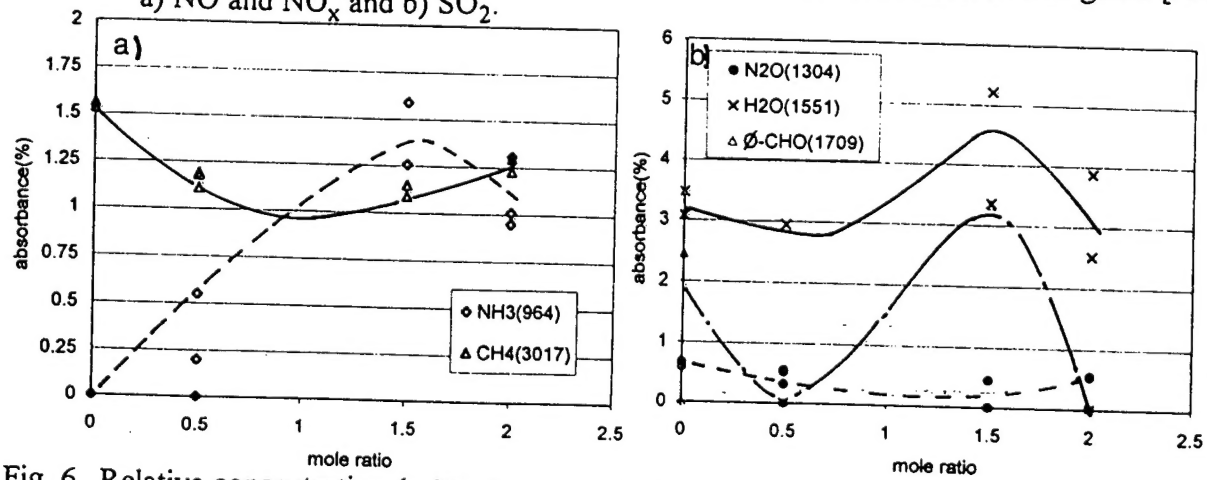

Fig. 6 Relative concentration (∝ IR absorbance) of a) NH_3 and CH_4 and b) N_2O, H_2O and ϕ-CHO (aldehyde) as a function of ammonia to acid gas molecule ratio at flue gas flow rate Q_f = 6 Nm^3/h (100 L/min), V =23 kV, $[NO]_o$ = 100 ppm and V_{add} = 5 m/s.

964, 3017, 1304, 1551 and 1709 cm^{-1} in Fig. 6. For the molecule ratio below 1, NO and NO_x are removed by a reduction reaction of methane and ammonia radicals, hence, removal rate of NO, NO_x, N_2O and CH_4 increase with increasing molecule ratio. However, for the molecule ratio larger than 1, ammonia and their radicals may be mainly consumed for SO_2 removal and ϕ-CHO formations hence NO, NO_x, N_2O and CH_4 removals may be reduced.

By comparison with combustion generated flue gas (without large methane concentration), the removal rate of NO_x is much lower as has been observed for the other engine generated flue gas plasma treatments [1-4] in spite of effective NO oxidations. The role played by the hydrocarbon under the non-thermal plasma should be investigated in detail.

CONCLUDING REMARKS

An experimental investigation has been conducted to remove acid gases from the stationary engine flue gas and the following concluding remarks are obtained: 1) NO, NO_x and SO_2 removal rates increase with increasing applied voltage and decrease with increasing NO initial concentration in the flue gases; (2) NO, NO_x and SO_2 removal rates non-monotonically depend on corona shower system additional gas flow velocity due to the mixing effects where the maximum removal rates are observed near 8 m/s; (3) SO_2 removal rate increases with increasing ammonia to acid gas molecule ratio while NO and NO_x non-monotonically depend on molecule ratio; (4) Ammonia and methane slips non-monotonically depend on ammonia to acid gas molecule ratio; (5) Trace unwanted discharge by-products significantly depend on ammonia to acid gas molecule ratio and only N_2O and ϕ-CHO (aldehydes) are observed; and (6) Significant amount of aerosol particles are formed during the acid gas removal processes.

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